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ORIGINAL ARTICLE

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Measured exposures by personal monitoring for respirable suspended particles and environmental tobacco smoke of housewives and office workers resident in Bremen, Germany

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Abstract *Objective*: Exposures to respirable suspended particles (RSP) and environmental tobacco smoke (ETS) were assessed in Bremen, Germany, as part of a European air quality study. The range and level of personal exposures were assessed for housewives and office workers. *Design*: Nonsmokers were randomly selected from a representative sample of the population of Bremen. Housewives were recruited into one group primarily for assessment of exposures in the home and office workers, into a second group for assessment of the contribution of the workplace to overall exposure. *Methods*: A total of 190 subjects collected air samples from areas close to their breathing zone by wearing personal monitors for 24 h. Samples collected were analysed for RSP, ultraviolet-absorbing particulate matter (UVPM), fluorescing particulate matter (FPM), solanesol-related particulate matter (SolPM), nicotine and 3-ethenylpyridine (3-EP). Saliva cotinine levels for all subjects were also established. *Results*: Overall the levels found were quite low, with the majority of results being below the limit of quantification. Workers both living and working with smokers were exposed to the highest 24-h median quantities of RSP (789 μg) and ETS particles (128 μg) measured by FPM. The highest nicotine levels, based on median 24-h time-weighted average concentrations, were experienced by office workers working with smokers (0.69 μg m^{-3}). These workers were also found to have the highest median cotinine levels (1.6 ng ml^{-1}). *Conclusions*: The most highly exposed workers, both living and working with smokers, would potentially inhale over 20 cigarette equivalents (CE) per annum as based on the upper decile levels. Housewives living with smokers could inhale up to 11 CE per annum as based on the upper decile levels. Locations outside the workplace, including the home, contribute most to overall RSP and ETS particle exposure. Consideration should be given to extending the personal monitoring period in cities where levels appear to be quite low.

Key words Personal exposure · Respirable suspended particles · Environmental tobacco smoke · Nicotine · Cotinine

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Introduction

The impact of air pollution on health is under detailed investigation in both Europe and the United States (US). An epidemiology study has commenced in the US to investigate air pollution and its impact in over 80 cities. This National Morbidity and Mortality Air Pollution Study (NMMAPS) is under the control of the Health Effects Institute, which is funded by industry and the United States Environmental Protection Agency (USEPA). The NMMAPS study will run over 3 years and one of its objectives will be to compare ambient monitoring data with measurements of personal exposure.

Personal air monitoring was chosen for this study in preference to static or ambient measurements so as to represent more accurately personal exposures to selected pollutants. The study involved subjects monitoring the air over 24-h periods in Bremen, Germany, during May 1995. Environmental tobacco smoke (ETS) particles were estimated using ultraviolet-absorbing particulate matter (UVPM), fluorescing particulate matter (FPM) and solanesol-related particulate matter (SolPM). Vapour-phase ETS exposures were also assessed by simultaneous measurement of the concentrations of nicotine and 3-ethenylpyridine (3-EP). For evaluation of

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exposures to ETS and respirable suspended particles (RSP), households and workplaces were classified as smoking or nonsmoking. Subjects also provided saliva samples for cotinine analysis and self-reported activities using diaries and questionnaires. Similar methodologies have been used in other recent studies (Heavner et al. 1996; Jenkins et al. 1996; Sterling et al. 1996; Baek et al. 1997).

Bremen was the fifth successive European city studied by these authors with regard to air quality following investigations in Stockholm (Phillips et al. 1996), Barcelona (Phillips et al. 1997a), Turin (Phillips et al. 1997b) and Paris (Phillips et al. 1997c). Bremen is the capital city of Germany's smallest state and has a population of 683,000. About 30% of the work force is employed in industry and a major proportion, in car or component manufacture.

The study set out to assess the exposure of housewives and office workers to RSP and ETS particles by obtaining accurate measurements of ETS exposure levels. The information collated herein should provide some meaningful data to allow informed and objective debate on issues related to passive smoking and exposures to RSP overall. The main objectives of this study were (1) to recruit random nonsmoking subjects who were representative of the population of Bremen into six separate lifestyle "cells"; (2) to determine the range and degree of personal exposure of these cells of selected subjects to RSP and ETS constituents by means of personal air sampling over a 24-h period; and (3) to monitor separately the workplace and locations away from the workplace, including the home, so as to assess the contribution of the workplace to overall exposure to ETS and RSP.

Methods

Recruitment of subjects

Recruitment was performed by Teleperformance, a large opinion research bureau resident in Germany, from files created using previously described procedures based on the Mosaic geodemographic classification system (Phillips et al. 1996). The sample was chosen to be compliant with the following criteria:

1. All subjects were to be nonsmokers living within 15 km of the Bremen city centre.
2. Equal proportions were to be taken from the three age groups 20–34, 35–49 and 50–64 years.
3. Equal percentage distribution, according to Mosaic groups, as for the population living within 15 km of the city centre.
4. Subjects were to be distributed between six "cells" as indicated in Table 1; cells 3–6 were set aside for office workers.

Subjects were recruited using randomly selected telephone numbers and were screened to confirm their eligibility to participate in the study. Suitable volunteers were given an appointment to attend an information/training session organised at the Hotel Mercure Columbus in Bremen.

A subject's household was classified as "smoking" if a smoker of cigarettes or pipes/cigars was resident within the household and also normally smoked within communal areas of the household. The smoking status of a workplace was defined by the absence/presence of smoking co-workers within 30 m of the subject's work station. These definitions were chosen for best representation of "real world" situations and for consistency across the different cities under study, the attitudes of residents varying considerably from country to country. The regulations governing air quality in the workplace were also different in each country at the time the study was undertaken.

Table 1 Cell categorisation by home and workplace status (Bremen)

Cell	Study type	Smoking status		Planned number
		Household	Workplace	
1	Single monitor	Smoking	–	55
2	Single monitor	Nonsmoking	–	40
3	Dual monitor	Smoking	Smoking	45
4	Dual monitor	Smoking	Nonsmoking	30
5	Dual monitor	Nonsmoking	Smoking	40
6	Dual monitor	Nonsmoking	Nonsmoking	30

Monitoring session

Subjects were required to wear a personal monitor designed to collect particulate and vapour-phase components present in the air close to the subject's breathing zone (Ogden et al. 1996). RSP and ETS particles were collected onto a Fluoropore membrane filter; nicotine and 3-EP were adsorbed onto XAD-4 resin beads. Each personal monitor was calibrated prior to issue and the calibrated flows were rechecked upon their return to ensure consistent sampling had been performed throughout the monitoring period. Data generated by personal monitors found to be out of calibration on their return, possibly resulting from tampering, misuse or equipment malfunction, were excluded. The personal monitoring methodology has been described in detail elsewhere (Phillips et al. 1996) and consisted in brief of the following.

Initial visit to the study centre

On arrival, subjects were shown an instructional video, dubbed into German, explaining the objectives of the air quality survey and were given further instructions regarding use of the equipment and how to complete the documentation by locally recruited interpreters. Subjects were issued German-language questionnaires and diaries for recording of exposures and observations over the 24-h collection period and were supervised during completion of a "first-visit" questionnaire. To avoid misinterpretation and possible errors in translation, all questionnaires and diaries were designed for either numeric or tick-box answers. Nonworking subjects recruited for participation in cells 1 and 2 were provided with a single personal monitor for use over the collection period (single monitor study). Working subjects recruited for participation in cells 3–6 were provided with two personal monitors for use over the same period (dual monitor study). All subjects were required to provide a saliva sample prior to the monitoring period (pre-sample).

Final visit to the study centre

Following completion of the 24-h monitoring period, subjects returned their personal monitors and associated documentation to the study centre. Subjects also provided a second saliva sample (post-sample) and completed a "last-visit" questionnaire.

Analytical procedures

All analytical procedures were validated and have been fully described elsewhere by these authors (Phillips et al. 1996). In this

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study the following analytes were determined: (1) *RSP* – using a gravimetric procedure (Ogden et al. 1990); (2) *saliva cotinine* – using a radioimmunoassay procedure (Van Vunakis et al. 1987; Davis and Stiles 1993); (3) *nicotine and 3-EP* – using a capillary gas chromatography (GC) procedure with nitrogen-specific detection (Ogden et al. 1989); and (4) *estimation of ETS particles (3 procedures)* – using high-performance liquid chromatography (HPLC) procedures to determine the ultraviolet absorbance (UVPM) or the fluorescence (FPM) or solanesol content (SolPM) of methanolic filter extracts (Ogden et al. 1990; Phillips et al. 1996); the factors used in this study to convert instrument responses into an equivalent concentration of ETS particles were 41 (SolPM), 43 (FPM) and 7.8 (UVPM) as determined by Nelson et al. (1997).

The analytical limits of quantification (LOQ) for all analytes are presented in Table 2 together with estimates for the corresponding air concentrations. For calculation of summary statistics, any data below the analytical LOQ were assigned a value of 1/2LOQ prior to the calculation of air concentrations. The analyte LOQs in air varied according to the sampling time and pump flow rate and were therefore different for each sample.

Subject selection

Of the 200 subjects who were initially recruited for the study, 5 were excluded because they failed to collect their samples and a further 5 subjects were excluded because their saliva cotinine levels were above 25 ng ml^{-1}. This threshold, to differentiate nonsmokers from smokers, was previously selected by these authors (Phillips et al. 1994) on the basis of Etzel's (1990) comprehensive review of saliva cotinine studies and has been discussed elsewhere (Phillips et al. 1997b).

The age and sex distributions of the remaining 190 subjects who successfully completed the study are presented in Table 3. This shows that the study distributions did not meet that planned of 50% per sex in the dual monitor study or 100% female in the single monitor study. There was a slight over-representation of the older age groups from the planned 33% in each group.

The 11 Mosaic groups represented by the German population are listed in Table 4, these groups being created by coarse classification of the 37 Mosaic types that have been identified in Germany. The overall distribution of these groups in Bremen and that of the subjects recruited for this study are depicted in Fig. 1. The more affluent groups were over-represented, possibly due to recruitment of office workers, a high percentage of whom were assumed to come from engineering companies. The participants were questioned about their occupation on the first-visit questionnaire and were restricted to a choice of 12 occupations from which to select and provide their answers. The occupations of the 107 subjects who responded to this question are listed in Table 5.

Pollutant information

Detailed information about ambient air quality during the course of the study was obtained locally (Senator für Umweltschutz und Stadtentwicklung). The levels quoted for outside ambient air concentrations can be used as a guide and, in the case of particulates, as a comparison with indoor levels. Concentrations of particulates, NO, NO_2, SO_2, O_3 and CO were obtained from four monitoring stations situated around the Bremen area. Overall daily mean NO_2 concentrations varied from 50 to 234 μg m^{-3} and were below 100 μg m^{-3} on only 4 days of the 26-day study period. The United Kingdom Department of the Environment pollutant bandings indicate that the air quality could be described as "poor" for the

Table 2 Limits of quantification for the analytical methods and corresponding air concentrations according to collection period (Bremen)

Measurement	Analytical LOQ	Corresponding LOQs in air (μg m^{-3})		
		24 h	14.99 h[a]	7.47 h[b]
Respirable suspended particles (RSP)[c]	24.5 μg/filter	9.90	15.9	31.8
ETS particles measured by UV (UVPM)[c]	1.17 μg/filter	0.47	0.76	1.52
ETS particles measured by fluorescence (FPM)[c]	0.28 μg/filter	0.11	0.18	0.36
ETS particles measured by solanesol (SolPM)[c]	0.62 μg/filter	0.25	0.40	0.80
Nicotine[d]	0.1 μg/tube	0.09	0.14	0.28
3-Ethenylpyridine (3-EP)[d]	0.1 μg/tube	0.09	0.14	0.28
Saliva cotinine	1.00 ng ml^{-1}	–	–	–

[a] Mean time spent outside the workplace for working subjects in Bremen
[b] Mean time spent at work for working subjects in Bremen
[c] Calculated assuming a flow rate of 1.72 l min^{-1} through the Fluoropore filter
[d] Calculated assuming a flow rate of 0.80 l min^{-1} through the XAD-4 tube

Table 3 Age and sex distribution for study subjects (Bremen) (*SH* smoking household, *NSH* nonsmoking household, *SW* smoking workplace, *NSW* nonsmoking workplace)

Cell	Sex			Age range (years)			Overall total
	Unknown	Males	Females	20–34	35–49	50–64	
1 (SH)		2	19	2	10	9	21
2 (NSH)		13	47	12	20	28	60
3 (SH, SW)		10	8	7	8	3	18
4 (SH, NSW)		4	2	3	3	0	6
5 (NSH, SW)	1	32	16	8	24	17	49
6 (NSH, NSW)	1	25	10	9	20	7	36
Single monitor total	0	15	66	21	30	37	81
Dual monitor total	1	72	36	27	55	27	109
Overall total	2	86	102	48	85	64	190

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Table 4 Mosaic group national distribution (Germany)

Mosaic group	Descriptor	National distribution
A	Luxury flats	4.4%
B	High-rise buildings	2.8%
C	Low-income urban areas	13.5%
D	Lower/middle-income urban areas	17.6%
E	Comfortable conservatives	9.0%
F	Average areas	3.4%
G	Up-and-coming suburbs	4.2%
H	Affluent communities	12.9%
I	Comfortable small-town dwellers	7.9%
J	Provincial communities	14.3%
K	Classic rural	10.0%

Table 5 Occupations of recruited subjects (Bremen)

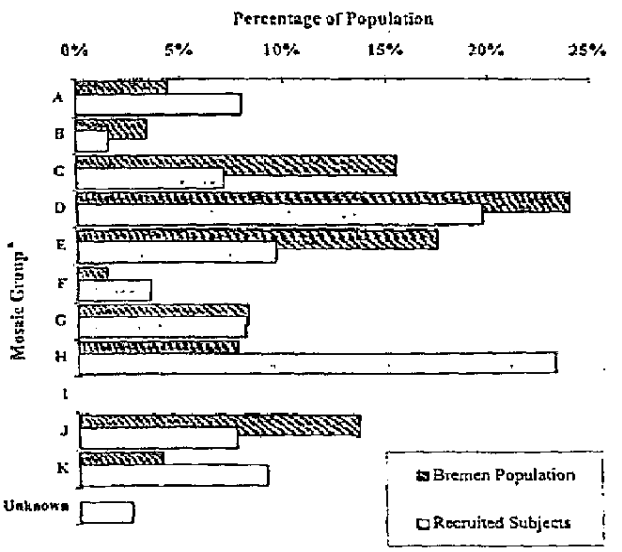
Occupation	Number of responses
Administrative/secretarial	17
Education	11
Engineering	15
Government agency (civil service)	13
Legal/financial (e.g. solicitor, banker)	8
Medical (e.g. doctor, nurse)	1
Other	20
Wholesale/retail (e.g. shop assistant)	13
Science/computing	8
Supply industry	1
Total	107

majority of the study period on the basis of NO_2 levels. These were the highest found to date in any of the European cities studied by these authors. The SO_2 levels were lower than those found in either Barcelona or Turin, with the overall mean value being 23 μg m^{-3} (min. 4.2 μg m^{-3}, max. 39 μg m^{-3}). The particulate concentrations were also quite low, the range being 11–58 μg m^{-3} and the overall mean value being 23 μg m^{-3}.

Results and discussion

For the assignment of a subject to a particular cell, procedures must be defined to classify the household and/or workplace as smoking or nonsmoking. In a recent US study, using a protocol similar to that applied in these European studies, Jenkins et al. (1996) reported data based on two separate procedures. Initially, subjects were assigned to a cell on the basis of their responses to the telephone screening questionnaire; cell categorisations were subsequently refined by rejection of subjects whose diary observations did not correspond to their initial cell assignments.

In this study, neither of the above-mentioned procedures were utilised; instead, cells were categorised according to the answers provided on the first-visit questionnaire. It was believed that responses to the screening questionnaire could not always be guaranteed due to variations in translations and to the possible incorrect categorisation of cells by telephonists working for the recruitment agencies. It was noted on the pump survey questionnaires that 36% of the subjects admitted to not fully completing their diaries; thus, the use of diary observations to refine cell categorisation was not considered.



* Mosaic group descriptors are listed in Table 4.

Fig. 1 Distribution of subjects' lifestyles (Bremen)

Comparison of measures for estimating ETS concentrations

There was an excellent correlation between ETS particle estimates using UVPM and FPM methods (R^2 = 0.98), with moderate correlations being found between SolPM and UVPM (R^2 = 0.71) and SolPM and FPM (R^2 = 0.78) estimates. The gradient of best-fit lines suggested the expected overall trend of UVPM > FPM > SolPM, an observation confirmed by Fig. 2; however, there was also an indication that SolPM may give higher estimates for ETS particles than FPM above approximately 20 μg m^{-3}. Also evident in Fig. 2 was the high number of SolPM results (71%) below the LOQ, resulting in "plateaux" on the distribution curve at the LOQs of the single and dual monitor studies; in comparison, only 3% of FPM findings and 5% of UVPM results were below the LOQ. In addition, of the 208 individual SolPM estimates below the LOQ, more than 25% and 15% of corresponding nicotine and 3-EP concentrations, respectively, were quantifiable.

Throughout this publication, ETS particle concentrations, corresponding cigarette equivalent (CE) calculations and comparisons between subject groups and

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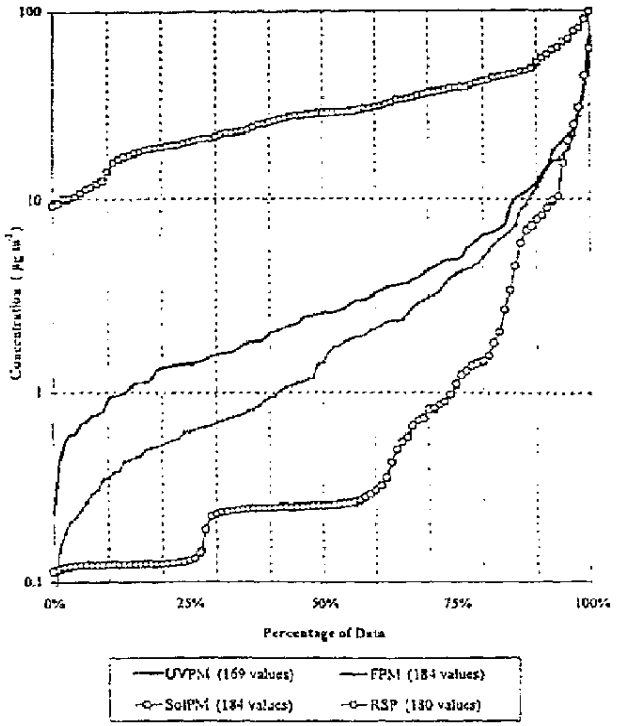

Fig. 2 Cumulative frequency distributions of 24-h TWA particle concentrations (Bremen)

cells have been based upon both FPM and SolPM determinations. It has previously been suggested (Ogden et al. 1990) that SolPM and, to some extent, FPM methods are more specific to ETS particles than the use of UVPM methods, which will be sensitive to other combustion sources. It was evident from the subjects' questionnaires that 86% of subjects believed their exposure to smoke sources other than tobacco was either "none" or "very low". Similar subjective assessments for ETS exposure were made by 83% of subjects.

There was a moderate correlation between nicotine and 3-EP measurements ($R^2 = 0.78$), with a gradient suggesting that nicotine concentrations were approximately 3 times higher than 3-EP levels. Neither vapour-phase analyte correlated very well with any of the particulate matter estimates ($R^2 = 0.42$–0.55). In this study, 68% and 62% of determined 3-EP and nicotine concentrations, respectively, were below the LOQ. The poor correlations observed in this study were believed to be due to the large proportion of results close to or below the LOQ, where acceptable assay reliability was at a minimum.

There were also poor correlations ($R^2 = 0.01$–0.30) for post-cotinine concentrations with both vapour- and particulate-phase ETS estimates. The distributions depicted in Fig. 3 also suggest very few similarities between the cumulative distributions of nicotine/3-EP and cotinine. In this study the method used for cotinine determination was initially selected only for the purpose of distinguishing between smokers and nonsmokers. More than 67% of determinations were not quantifiable using the current LOQ of 1 ng ml^{-1}; therefore, the use of cotinine measurements as a marker for ETS exposure was not considered reliable in this instance.

These authors are aware of more recent methods for cotinine analysis and are currently validating an assay for cotinine in saliva below 0.1 ng ml^{-1}. Once a highly sensitive assay has been established, cotinine may prove to be the most reliable biomarker for assessment of ETS exposure. However, the use of much lower limits of quantification will require that more attention be paid to such considerations as dietary nicotine contributions and concentration variations attributable to the method of sample collection (Schneider et al. 1997).

Concentrations of ETS constituents to which Bremen subjects were exposed

In this publication, median values have primarily been used for reporting RSP and ETS marker concentrations because the data generated were not normally distributed. Arithmetic and geometric means for each data set have been also been reported together with 10th and 90th percentile values (lower and upper deciles) as an

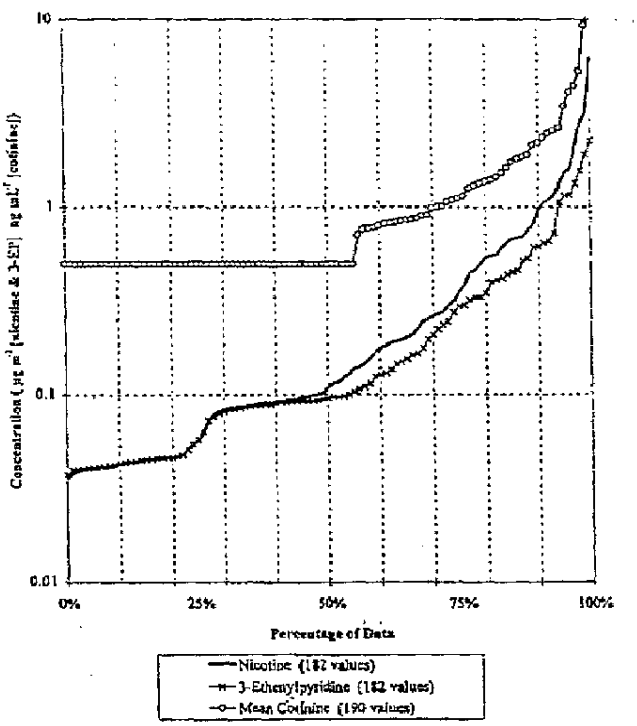

Fig. 3 Cumulative frequency distributions of nicotine, 3-EP and cotinine (Bremen)

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indication of the range of values more appropriate than the minimum and maximum.

Particulate and vapour-phase components measured for housewives were compared by cell with calculated time-weighted average (TWA) concentrations recorded for individual workers. These calculations were based on measured concentrations and the operational time over which the monitors were used inside and outside the workplace. These data are summarised in Tables 6 and 7, with corresponding cumulative frequency distributions for ETS particles and nicotine being shown in Figs. 4–6. The significance of any concentration differences between cells was examined using the Wilcoxon rank-sum test. Prior to the application of this nonparametric test, Kruskal-Wallis nonparametric analysis of variance (ANOVA) was applied to the data so as to detect if there was an overall difference between the cells. If the overall Kruskal-Wallis analysis proved nonsignificant ($P > 0.05$), any significance detected using the Wilcoxon rank-sum test would be considered as false positive. For all the analytes investigated, the Kruskal-Wallis ANOVA provided evidence of a significant overall difference between cells, and subsequent pairwise comparisons of cells were performed using the Wilcoxon rank-sum test.

As shown in Table 6, the median SolPM concentrations recorded for cells 1 and 2, housewives from smoking households (SH) and nonsmoking households (NSH), respectively, were equivalent. This finding was

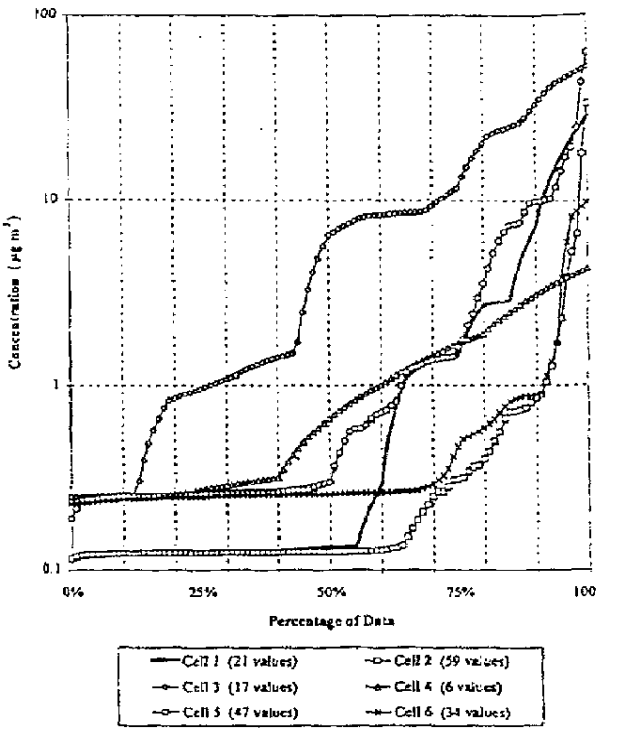

Fig. 4 Cumulative frequency distributions of SolPM by cell (Bremer

Table 6 Summary of the statistics for 24-h TWA particle concentrations recorded for all subjects[a] (Bremen) (*SH* smoking household *NSH* nonsmoking household, *SW* smoking workplace, *NSW* nonsmoking workplace)

Analyte	Cell	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
RSP ($\mu g\ m^{-3}$)	1 (SH)	21	21	63	39	35	36
	2 (NSH)	58	14	37	27	25	25
	3 (SH, SW)	17	24	86	48	42	39
	4 (SH, NSW)	6	19	49	35	31	36
	5 (NSH, SW)	45	14	51	32	29	29
	6 (NSH, NSW)	33	12	38	25	23	23
SolPM ($\mu g\ m^{-3}$)	1 (SH)	21	0.12	7.3	3.1	0.49	0.13
	2 (NSH)	59	0.12	0.84	1.0	0.23	0.12
	3 (SH, SW)	17	0.25	33	11	3.5	6.5
	4 (SH, NSW)	6	0.24	3.1	1.3	0.73	0.66
	5 (NSH, SW)	47	0.25	9.9	3.8	0.83	0.30
	6 (NSH, NSW)	34	0.24	0.87	0.89	0.39	0.26
FPM ($\mu g\ m^{-3}$)	1 (SH)	21	0.46	17	6.1	2.8	3.4
	2 (NSH)	59	0.21	3.0	1.6	0.82	0.75
	3 (SH, SW)	17	1.8	36	14	6.3	6.3
	4 (SH, NSW)	6	1.0	6.0	3.3	2.4	2.8
	5 (NSH, SW)	47	0.79	13	5.6	2.7	3.3
	6 (NSH, NSW)	34	0.38	3.2	1.5	0.96	0.65
UVPM ($\mu g\ m^{-3}$)	1 (SH)	20	1.0	16	6.6	4.0	4.2
	2 (NSH)	59	0.61	4.5	2.6	1.6	1.4
	3 (SH, SW)	14	3.4	41	18	12	9.7
	4 (SH, NSW)	4	3.2	8.8	5.5	4.8	4.1
	5 (NSH, SW)	41	1.6	14	6.3	4.1	3.6
	6 (NSH, NSW)	31	1.3	4.5	2.8	2.2	1.7

[a] TWA exposure concentrations, determined for each subject from measured levels both inside and outside the workplace, were used to calculate the statistical parameters for cells 3–6

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Table 7 Summary of the statistics for cotinine and 24-h TWA nicotine and 3-EP concentrations recorded for all subjects[a] (Bremen) (*SH* Smoking household, *NSH* nonsmoking household, *SW* smoking workplace, *NSW* nonsmoking workplace, *3-EP* 3-ethenlypyridine)

Analyte	Cell	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
Nicotine ($\mu g\ m^{-3}$)	1 (SH)	21	0.04	1.5	0.63	0.31	0.49
	2 (NSH)	59	0.04	0.22	0.10	0.07	0.05
	3 (SH, SW)	17	0.13	2.1	1.2	0.66	0.69
	4 (SH, NSW)	6	0.09	0.63	0.32	0.23	0.23
	5 (NSH, SW)	44	0.09	1.1	0.52	0.26	0.20
	6 (NSH, NSW)	35	0.08	0.24	0.16	0.13	0.10
3-EP ($\mu g\ m^{-3}$)	1 (SH)	21	0.04	1.5	0.54	0.27	0.32
	2 (NSH)	59	0.04	0.13	0.07	0.06	0.05
	3 (SH, SW)	17	0.14	1.3	0.69	0.48	0.61
	4 (SH, NSW)	6	0.09	0.53	0.28	0.22	0.24
	5 (NSH, SW)	44	0.09	0.53	0.29	0.20	0.15
	6 (NSH, NSW)	35	0.08	0.25	0.13	0.11	0.09
Cotinine[b] ($ng\ ml^{-1}$)	1 (SH)	21	0.50	2.4	1.6	1.2	1.4
	2 (NSH)	60	0.50	1.3	1.1	0.70	0.50
	3 (SH, SW)	18	0.50	3.1	2.1	1.5	1.6
	4 (SH, NSW)	6	0.50	3.4	1.5	0.85	0.50
	5 (NSH, SW)	49	0.50	1.6	1.0	0.80	0.50
	6 (NSH, NSW)	36	0.50	0.88	0.66	0.61	0.50

[a] TWA exposure concentrations, determined for each subject from measured levels both inside and outside the workplace, were used to calculate the vapour-phase statistical parameters for cells 3–6

[b] Values calculated from the average of pre- and post-monitoring saliva cotinine concentrations

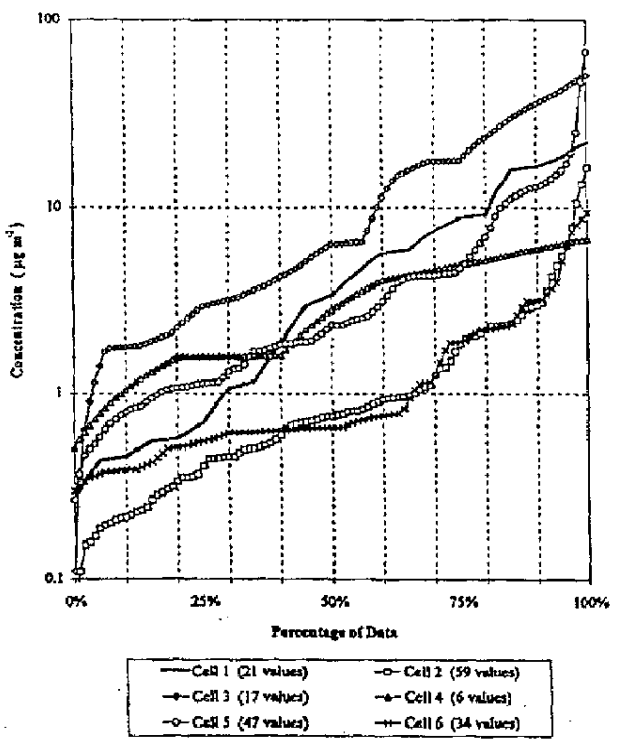

Fig. 5 Cumulative frequency distributions of FPM by cell (Bremen)

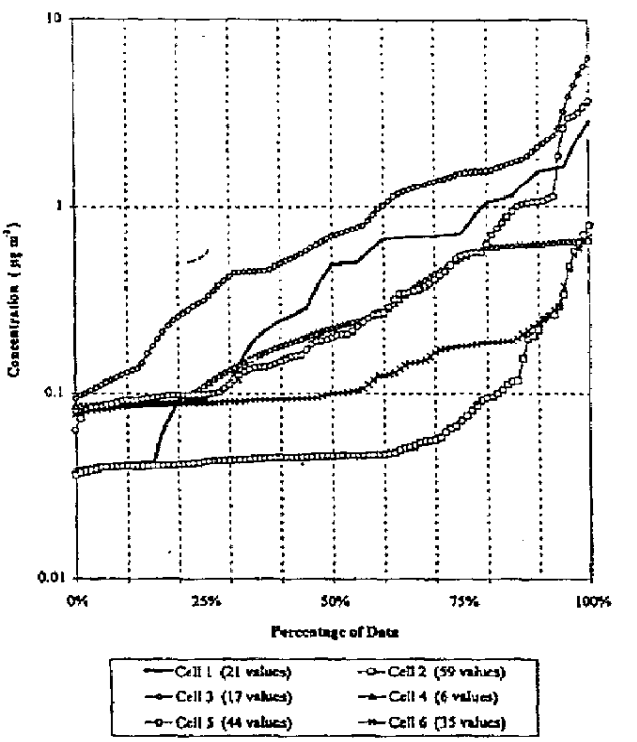

Fig. 6 Cumulative frequency distributions of nicotine by cell (Bremen)

supported by the Wilcoxon rank-sum test, which did not indicate any significant difference ($P > 0.05$) between these cells. However, examination of the 90th percentile data suggested that subjects from cell 1 were exposed to levels more than 8 times higher than those from cell 2. These observations could be explained by

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examination of Fig. 4, which showed that more than 50% of the data were below the LOQ; hence, the cells had equivalent medians. Also evident from Fig. 4 were the higher levels recorded for cell 1 from approximately the 60th percentile upwards. These observations highlight the need to examine all data, not just medians, and show that Wilcoxon rank-sum test results could be misleading.

Conversely, the median SolPM level recorded for cell 6, workers from NSH and nonsmoking workplaces (NSW), was more than twice that noted for cell 2 (housewives from NSH). The Wilcoxon rank-sum test also showed a highly significant difference ($P \leq 0.001$) between these cells, whilst the 90th percentile levels were similar. Again, examination of Fig. 4 showed that most of these data were below the LOQ, the differences in LOQs for these cells resulting in apparently higher levels for cell 6. The distribution curves were very similar for these cells above the 75th percentile, where levels of airborne solanesol were quantifiable.

The above mentioned examples show that where the majority of data are close to or below the LOQ, extreme caution should be taken in comparing exposures between cells on the basis of either the median concentrations or statistical ranking. It also suggests that values other than the median may be a more appropriate measure for the comparison of exposures between cells. The 90th percentile values can provide more meaningful comparisons; however, care is required to ensure that a sufficient number of subjects exist within the cells to avoid bias from isolated high values.

It would appear that workers living in smoking households and working in smoking workplaces (cell 3) were the most highly exposed group overall, with the median RSP concentration being 39 $\mu g\ m^{-3}$ and the ETS particle contribution being 6.5 $\mu g\ m^{-3}$ (16.8%) as based on SolPM and 6.3 $\mu g\ m^{-3}$ (16.2%) as based on FPM measurements. In a recent US study covering 16 cities and almost 1500 subjects, Jenkins et al. (1996) found 24-h TWA concentrations of 34 $\mu g\ m^{-3}$ RSP, 3.8 $\mu g\ m^{-3}$ ETS particles as based on SolPM and 8.6 $\mu g\ m^{-3}$ ETS particles as based on FPM for subjects living and working with smokers. If we compare the median concentrations to which workers from nonsmoking homes and workplaces were exposed in this study with the US study (Jenkins et al. 1996), then median RSP concentrations were 23 $\mu g\ m^{-3}$ (15.2 $\mu g\ m^{-3}$), levels of ETS particles as based on SolPM were 0.26 $\mu g\ m^{-3}$ (0.09 $\mu g\ m^{-3}$ or half the LOQ) and levels of ETS particles as based on FPM were 0.65 $\mu g\ m^{-3}$ (0.665 $\mu g\ m^{-3}$). From these results, only measurements using SolPM to estimate ETS particles in this study differed appreciably from those recorded in the US study.

In another study recently conducted in Munich, Germany, Scherer et al. (1995) reported comparable median levels of nicotine for nonsmokers (0.07 and 0.30 $\mu g\ m^{-3}$ from nonsmoking and smoking households, respectively). Their study comprised a 7-day monitoring period using passive samplers designed specifically for the collection of nicotine and 3-EP (Ogden and Maiolo 1992). Their median levels of 3-EP were considerably lower than those evident in this study, possibly due to the low response of passive samplers to 3-EP.

Exposures to RSP, ETS particles and nicotine

The term *exposure* is often used in defining maximal allowable concentrations for hazardous compounds and is normally determined by fixed-site monitoring over standard periods. In the context of this personal monitoring study, where concentrations cannot be directly related to a specific environment, the term *exposure* is used as a measure of "potential inhaled quantity" and was calculated as the product of the analyte concentration, the time subjected to such concentration and the breathing rate maintained throughout the period. A similar assumption was recently made by Ogden and Martin (1997), who noted that this provided "a more accurate accounting of total exposure among individuals as they engage in different activities in different microenvironments". Where exposures have been quoted in terms of CE, these have been calculated in relation to the mainstream particle (tar) and nicotine yields of typical German cigarettes, although it is recognised that the particle phases of ETS and mainstream smoke differ considerably in composition and particle size. The values, 11 mg ETS particles and 0.8 mg nicotine, were calculated from the mean yields of the top six selling cigarette-brand types in Germany. In this publication, CEs are used solely for conceptual comparison of exposures between groups of nonsmokers. The factor (Ogden and Martin 1997) used to relate exposure of nonsmokers to that of smokers was not applied to the data.

Daily exposures in terms of potential inhaled amounts (micrograms) calculated for each cell over the 24-h monitoring period are summarised in Table 8. A breathing rate of 0.65 $m^3\ h^{-1}$, the average level of respiration calculated for "awake" females, was used for calculating housewife exposures in cells 1 and 2. For cells 3–6, where exposures were not sex-dependent, a breathing rate of 0.85 $m^3\ h^{-1}$ was used, this being an average of the breathing rates for "awake" males (1.05 $m^3\ h^{-1}$) and females (0.65 $m^3\ h^{-1}$) as reported by Holcomb (1993). A comparable average breathing rate of 0.93 $m^3\ h^{-1}$ has recently been used by Jenkins et al. (1996) to estimate exposures on the 16-city US study using personal monitoring methods similar to those used in this study. Both median and 90th percentile values have been calculated to represent "typical" and "highly exposed" subjects, respectively.

For estimation of annual exposures, separate procedures were adopted for housewives and for workers. The daily exposures calculated for housewives were multiplied by 365 to obtain an estimate of annual exposure. For cells 3–6 (workers) the median and 90th percentile levels were calculated from data provided by the indi-

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Table 8 Calculated 24-h exposures to RSP, ETS particles and nicotine using median and 90th percentile levels[a] (Bremen) (*SH* smoking household, *NSH* nonsmoking household, *SW* smoking workplace, *NSW* nonsmoking workplace)

Cell	RSP (μg)	ETS particles		
		SolPM (μg)	FPM (μg)	Nicotine (μg)
Median levels:				
1 (SH)	561	2.0	53	7.6
2 (NSH)	389	1.9	12	0.78
3 (SH, SW)	789	132	128	14
4 (SH, NSW)	732	13	58	4.7
5 (NSH, SW)	584	6.1	48	4.1
6 (NSH, NSW)	466	5.3	13	2.0
90th percentiles:				
1 (SH)	976	114	259	24
2 (NSH)	575	13	47	3.4
3 (SH, SW)	1760	680	726	42
4 (SH, NSW)	1001	64	122	13
5 (NSH, SW)	1034	202	263	22
6 (NSH, NSW)	784	18	65	4.9

[a] A breathing rate of 0.65 $m^3 h^{-1}$ was assumed for housewives (cells 1, 2) and 0.85 $m^3 h^{-1}$, for working subjects (cells 3–6)

vidual monitors worn in the workplace and away from the workplace. Working subjects were assumed to have a breathing rate of 0.85 $m^3 h^{-1}$ at all times and to spend 35 h per week and 48 weeks per year in the workplace, with the remainder of the time being spent "at home". Annual exposure calculations for all subjects assumed no variation in ETS marker concentrations throughout the year, including weekends, from those measured during the monitoring period. Estimated annual exposures using the above assumptions and the median and upper decile concentrations are reported in Table 9 together with estimates for ETS particle and nicotine exposures in terms of CE.

As can be seen from Table 8, the highest daily levels of exposure to RSP, ETS particles and nicotine were recorded for workers living and working in smoking environments. The least exposed subjects over a 24-h period for all analytes were housewives from nonsmoking households.

If the exposures are annualised (Table 9) and cells are then ranked by median values for RSP levels, we find cell 3 (SH, SW) > cell 4 (SH, NSW) > cell 1 (SH) > cell 5 (NSH, SW) > cell 6 (NSH, NSW) > cell 2 (NSH). A similar trend is evident for ETS particles and nicotine, with cell 3 (workers from smoking homes and workplaces) being the most highly exposed and cell 2 (housewives from nonsmoking homes) being the least exposed. This would suggest that office workers overall are more highly exposed to RSP, ETS particles and nicotine than housewives. It is also apparent that the smoking status of a subject's home is a more significant factor in annual exposure than the smoking status of the workplace.

It is evident from Table 9 that exposures in this study were quite low, with no group being exposed to more than 5 CE per annum as based upon median levels. The most highly exposed group (90th percentile levels), nonsmokers in this study who worked and lived with smokers, would be exposed to less than 25 CE per annum. Conversely, if they lived and worked with nonsmokers they would be exposed to less than 3 CE per annum. The median annualised levels of exposure

Table 9 Estimated annual exposures for all subjects to RSP, ETS particles and nicotine[a] (Bremen) (*SH* smoking household, *NSH* nonsmoking household, *SW* smoking workplace *NSW* nonsmoking workplace)

Cell	Annual exposure (mg)						
	ETS particles				Cigarette equivalents		
	RSP	SolPM	FPM	Nicotine	SolPM	FPM	Nicotine
Median levels:							
1 (SH)	205	0.74	19	2.8	0.07	1.8	3.5
2 (NSH)	142	0.68	4.3	0.28	0.06	0.39	0.36
3 (SH, SW)	276	8.8	28	3.7	0.80	2.6	4.6
4 (SH, NSW)	271	3.1	13	1.2	0.28	1.2	1.5
5 (NSH, SW)	184	1.9	9.6	0.84	0.17	0.88	1.0
6 (NSH, NSW)	154	1.8	4.6	0.65	0.16	0.42	0.81
90th percentiles:							
1 (SH)	356	42	95	8.8	3.8	8.6	11
2 (NSH)	210	4.8	17	1.3	0.43	1.5	1.6
3 (SH, SW)	704	269	252	17	24	23	22
4 (SH, NSW)	467	35	63	6.3	3.2	5.7	7.9
5 (NSH, SW)	382	36	58	5.1	3.3	5.3	6.4
6 (NSH, NSW)	302	9.7	27	2.3	0.88	2.4	2.9

[a] A breathing rate of 0.65 $m^3 h^{-1}$ was assumed for housewives (cells 1, 2) and 0.85 $m^3 h^{-1}$, for working subjects (cells 3–6). Annual exposures for housewives were calculated by simple extrapolation of their 24-h exposure levels. Exposure calculations for working subjects were made by assuming a 35-h working week and a 48-week working year, with the remainder of the time being spent "at home". Concentrations of ETS markers noted for workers at "work" and at "home" were taken from the data recorded for the individual monitors of each cell

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(4.6 CE) noted for office workers living and working with smokers in Bremen are 5 times higher than those found by these authors in Stockholm (0.9 CE) and approximately half those found in Barcelona (11.9 CE) and Turin (9.9 CE).

It was also possible by applying the same criteria used for the calculations in Table 9 to estimate the contribution of the workplace to overall annual exposure. These estimates, expressed as a percentage of both median and upper decile results, are summarised in Table 10 and show that the contributions vary from approximately 20% in cell 4 (SH, NSW) to over 50% in cell 5 (NSH, SW). The estimates based on the upper decile levels probably provide the most accurate assessment of overall workplace contribution as most of the median levels are close to or below the LOQ. If the results from the individual monitors for all workers are pooled, the workplace contributes between 35% and 37% of annual exposure to both ETS particles (based upon FPM) and nicotine, irrespective of whether median or upper decile levels are used in the calculation.

Table 10 Estimated contribution of the workplace to annual exposures to RSP, ETS particles and nicotine recorded for all subjects (Bremen) (*SH* smoking household, *NSH* nonsmoking household, *SW* smoking workplace, *NSW* nonsmoking workplace)

Cell	RSP	ETS particles		Nicotine
		SolPM	FPM	
Median levels:				
3 (SH, SW)	20%	11%	16%	23%
4 (SH, NSW)	16%	63%	26%	23%
5 (NSH, SW)	28%	35%	36%	43%
6 (NSH, NSW)	17%	32%	31%	35%
90th percentiles:				
3 (SH, SW)	19%	10%	18%	39%
4 (SH, NSW)	19%	14%	15%	14%
5 (NSH, SW)	38%	86%	67%	65%
6 (NSH, NSW)	24%	14%	19%	22%

Concentrations of RSP, ETS particles, nicotine and 3-EP as based on location

The magnitude of exposures to RSP, ETS particles and nicotine for working subjects both inside and outside of the workplace were assessed using data provided by the individual monitors. Individual monitor contributions were combined to provide an estimate of exposure concentrations in smoking (Table 11) and nonsmoking (Table 12) environments both inside and outside the workplace. Comparison of saliva cotinine levels was not meaningful using this procedure and the data have been excluded from the tables.

As would be expected, median levels of RSP, ETS particles and nicotine were found to be higher in smoking environments than in nonsmoking environments. There is a remarkably close similarity between the median concentrations of analytes determined in the two smoking environments (Table 11), the major difference being noted between the vapour-phase analytes, nicotine and 3-EP. It is noteworthy that there are close similarities with the "home" data shown in Table 11 and the data recorded for cell 1 (housewives from SH) and presented in Tables 6 and 7, suggesting no difference between housewife and worker exposures outside the workplace.

Comparison of median levels in the two nonsmoking environments would initially suggest that the workplace has the highest levels; however, this conclusion would be based on data below the LOQ and examination of upper decile levels shows very little difference between the two environments. Consideration might be given in future studies to extension of the monitoring period for several days, even a whole week, so as to collect a larger sample, although subjects may not maintain study integrity over longer periods.

Table 11 Summary of the analytical statistics recorded for employed subjects in smoking environments (Bremen)

Analyte	Environment	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
RSP ($\mu g\ m^{-3}$)	Work[a]	63	15	105	46	36	37
	Home[b]	23	16	89	44	36	37
SolPM ($\mu g\ m^{-3}$)	Work[a]	65	0.35	22	7.9	1.3	0.59
	Home[b]	23	0.16	27	10	1.5	1.1
FPM ($\mu g\ m^{-3}$)	Work[a]	65	0.80	29	12	4.1	3.7
	Home[b]	23	0.62	23	11	3.7	3.7
UVPM ($\mu g\ m^{-3}$)	Work[a]	57	2.4	34	16	7.6	6.7
	Home[b]	18	1.6	25	15	6.7	5.8
Nicotine ($\mu g\ m^{-3}$)	Work[a]	63	0.12	3.3	1.3	0.44	0.32
	Home[b]	23	0.06	1.7	0.65	0.33	0.40
3-EP ($\mu g\ m^{-3}$)	Work[a]	63	0.12	1.7	0.63	0.34	0.25
	Home[b]	23	0.06	1.2	0.51	0.29	0.39

[a] Data from "workplace" monitor of subjects in cells 3 and 5
[b] Data from "home" monitor of subjects in cells 3 and 4

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Table 12 Summary of the analytical statistics recorded for employed subjects in nonsmoking environments (Bremen)

Analyte	Environment	Number of subjects	10th percentile	90th percentile	Arithmetic mean	Geometric mean	Median
RSP ($\mu g\ m^{-3}$)	Work[a]	40	14	53	28	24	21
	Home[b]	81	8.6	39	25	21	22
SolPM ($\mu g\ m^{-3}$)	Work[a]	40	0.32	1.7	0.89	0.55	0.42
	Home[b]	84	0.16	0.91	0.94	0.30	0.20
FPM ($\mu g\ m^{-3}$)	Work[a]	40	0.53	3.8	1.8	1.2	1.0
	Home[b]	84	0.32	3.6	1.6	0.80	0.67
UVPM ($\mu g\ m^{-3}$)	Work[a]	35	1.3	6.7	3.7	2.8	2.7
	Home[b]	76	0.95	4.5	2.5	1.8	1.5
Nicotine ($\mu g\ m^{-3}$)	Work[a]	42	0.11	0.45	0.22	0.19	0.16
	Home[b]	82	0.06	0.33	0.18	0.11	0.08
3-EP ($\mu g\ m^{-3}$)	Work[a]	42	0.11	0.33	0.20	0.17	0.15
	Home[b]	82	0.06	0.32	0.14	0.09	0.07

[a] Data from "workplace" monitor of subjects in cells 4 and 6
[b] Data from "home" monitor of subjects in cells 5 and 6

Subjective comparisons of ETS exposure

Observational diaries were maintained by subjects throughout the monitoring periods and a further last-visit survey questionnaire was completed at the end of the study. It is noteworthy that information from these sources indicated that approximately 42% of subjects working in smoking environments did not see or smell any evidence of smoking during the monitoring period. Also apparent from this information was the observation that about 21% of all subjects working in nonsmoking environments did note smoking during the monitoring period. These observations might indicate an incomplete segregation of smokers and nonsmokers and highlight the difficulties, as discussed previously, of defining workplaces as "smoking" or "nonsmoking". They do, however, represent "real world" situations and, as such, are meaningful.

Outside the workplace, using a combination of subjects on the single and dual monitor studies, approximately 18% of subjects from a smoking household did not note any smoking during the monitoring period, whereas 28% of subjects from a nonsmoking household noted smoking on their diaries or questionnaires.

As part of the last-visit survey, subjects were asked a number of subjective questions regarding their exposure to ETS, both in general and during the 24-h monitoring period. The environments regarded by subjects as their single most exposed location to ETS are listed in Table 13. This table shows that twice as many people believe the workplace, rather than the home, is where they receive their highest exposure to ETS. Also evident is the perception that restaurants/bars contribute most to ETS exposure. These observations suggest that subjects associate locations having high ETS concentrations with high exposure and do not take into account the amount of time spent in the location.

Conclusions

The fifth city completed by these authors in this series of personal monitoring studies was Bremen in northern Germany. The study was designed to recruit representative but randomly selected subjects from Bremen. However, the study attracted an over-representation of subjects from more affluent groups due to recruitment of more highly paid office workers from engineering companies located in the city.

Pollutant information from the local authorities indicated that during the study period, whilst overall particulate concentrations were low, levels of NO_2 were the highest encountered to date in any of the European cities monitored.

Office workers living and working with smokers were exposed to the highest 24-h TWA concentrations of RSP, ETS particles and nicotine in this study. The RSP levels found for these workers were 14% higher and the ETS particles as based on SolPM were 71% higher than those found in a recent 16-city study in the US. Conversely, using FPM measurements the ETS particle concentrations found in Bremen were 36% lower than those found in the US study. The determination of ETS particles found in this study resemble

Table 13 Subjective assessment of the environment where subjects consider themselves to be most exposed to ETS (Bremen)

Environment	% Responses[a]
Restaurants/bars	57
Work	12
In other types of buildings	5.8
Home	5.8
Nowhere/not exposed	2.6
Travelling/driving	2.2
Outdoors	1.1

[a] Responses were calculated as a percentage of total recruits; 27 subjects failed to answer this question correctly

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those of other studies most closely when FPM measurements are used.

Office workers living and working with smokers also potentially receive the highest annualised exposures to RSP and ETS particles. The lowest exposures were recorded for housewives living in nonsmoking households. As based on FPM median levels, the most highly exposed individuals would potentially receive approximately 6 times the amount of ETS particles than the least exposed subjects. This ratio increased to about 15:1 when the 90th percentile FPM levels were compared.

The home and environments outside the workplace contributed most to overall RSP and ETS particle exposures for the majority of subjects in this study. The levels encountered, however, were quite low and, similar to those found in Stockholm, the majority of the results were below the LOQ. The most highly exposed subjects to ETS particles would be subjected to approximately 22 CE/year, whilst the least exposed would receive less than 2 CE/year as based on upper decile levels.

In cities where levels appear to be quite low it may be more appropriate to sample over periods of up to 1 week. This extended period may give more accurate and representative sampling whilst improving analytical and statistical evaluation. Conversely, participants might modify their behaviour during an extended period and compromise study integrity.

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